EUR 477. e

EUROPEAN ATOMIC ENERGY COMMUNITY — EURATOM

HIGH PRECISION 4 $\pi\beta$-γ- COINCIDENCE COUNTING AND THE PREPARATION OF SOLID Co^{60} STANDARD SOURCES AND SOLUTIONS FOR AN INTERNATIONAL COMPARISON

by

A. SPERNOL, E. DE ROOST and O. LERCH

1964

Joint Nuclear Research Center
Geel Establishment — Belgium
Central Bureau for Nuclear Measurements (CBNM)

LEGAL NOTICE

This report can be obtained, at the price of Belgian Francs 50,– from : PRESSES ACADÉMIQUES EUROPÉENNES,
98, Chaussée de Charleroi, Brussels 6.
Please remit payments :

— to BANQUE DE LA SOCIÉTÉ GÉNÉRALE (Agence Ma Campagne) — Brussels — account No 964.558,

— to BELGIAN AMERICAN BANK AND TRUST COMPANY — New York — account No 121.86,

— to LLOYDS BANK (Foreign) Ltd. — 10 Moorgate — London E.C.2,

giving the reference : "EUR 477.e — High precision $4\pi\beta$-γ-coincidence counting and the preparation of solid Co^{60} standard sources and solutions for an international comparison."

Printed by Ceuterick, Louvain.
Brussels, February 1964.

EUR 477.e

HIGH PRECISION $4\pi\beta$-γ-COINCIDENCE COUNTING AND THE PREPARATION OF SOLID Co^{60} STANDARD SOURCES AND SOLUTIONS FOR AN INTERNATIONAL COMPARISON by A. SPERNOL, E. DE ROOST, and O. LERCH

European Atomic Energy Community — EURATOM
Joint Nuclear Research Center
Geel Establishment (Belgium)
Central Bureau for Nuclear Measurements (CBNM)
Brussels, February 1964 — pages 29 + figures 12.

The preparation and properties of electrolytically deposited Co^{60} sources and Co^{60} standard solutions are described. In addition the $4\pi\beta$-γ-coincidence method used for the determination of their activities is discussed in detail with particular reference to the corrections which have to be applied to the measured results and their control.

EUR 477.e

HIGH PRECISION $4\pi\beta$-γ COINCIDENCE COUNTING AND THE PREPARATION OF SOLID Co^{60} STANDARD SOURCES AND SOLUTIONS FOR AN INTERNATIONAL COMPARISON by A. SPERNOL, E. DE ROOST, and O. LERCH

European Atomic Energy Community — EURATOM
Joint Nuclear Research Center
Geel Establishment (Belgium)
Central Bureau for Nuclear Measurements (CBNM)
Brussels, February 1964 — pages 29 + figures 12.

The preparation and properties of electrolytically deposited Co^{60} sources and Co^{60} standard solutions are described. In addition the $4\pi\beta$-γ-coincidence method used for the determination of their activities is discussed in detail with particular reference to the corrections which have to be applied to the measured results and their control.

EUR 477. e

EUROPEAN ATOMIC ENERGY COMMUNITY — EURATOM

HIGH PRECISION 4 $\pi\beta$-γ- COINCIDENCE COUNTING AND THE PREPARATION OF SOLID Co^{60} STANDARD SOURCES AND SOLUTIONS FOR AN INTERNATIONAL COMPARISON

by

A. SPERNOL, E. DE ROOST and O. LERCH

1964

Joint Nuclear Research Center
Geel Establishment — Belgium

Central Bureau for Nuclear Measurements (CBNM)

CONTENTS

KEY TO THE FIGURES

KEY TO THE TABLES

HIGH-PRECISION $4\pi\beta$-γ-COINCIDENCE COUNTING AND THE PREPARATION OF SOLID Co^{60} STANDARD SOURCES AND SOLUTIONS FOR AN INTERNATIONAL COMPARISON

SUMMARY

The preparation and properties of electrolytically deposited Co^{60} sources and Co^{60} standard solutions are described. In addition the $4\pi\beta$-γ-coincidence method used for the determination of their activities is discussed in detail with particular reference to the corrections which have to be applied to the measured results and their control.

1 — INTRODUCTION

It has been well known since at least 1959, when Campion published his paper on $4\pi\beta$-γ-coincidence counting [1], that this method allows accuracies of the order of 0.1% in the measurement of radio-isotopes decay rates. This exceptionally high precision was the reason why BIPM proposed and organized an international comparison of solid Co^{60} sources in 1962/1963. CBNM who have carried out several investigations on the $4\pi\beta$-γ-method since 1959, were commissioned by BIPM to prepare and together with NPL to measure the sources needed.

In this paper the work of CBNM for BIPM distribution will be described, together with the method of measurement, which is based on older investigations. One of the chief aims of this report is to supply the participants in the BIPM distribution with all details of the source preparation, the source properties and the measurement of the sources which they had for comparison. A thorough critical discussion of the method and its practice is the other main purpose of this work.

2 — PREPARATION OF THE SOLID SOURCES

2.1 — Types of source, mounts, gold cover, containers

The method and organization of the BIPM distribution is described in a BIPM letter of 12th February 1963. According to this letter, 4 solid sources were to be prepared for each participating laboratory, 3 with a very high β-efficiency and activities of approximately 1000, 3000 and 5000 dps and one with a β-efficiency of approx. 50%. All sources had to be prepared on aluminium mounts 0.1 mm thick, 38 or 30 mm in outside diameter and 16 mm in inside diameter.

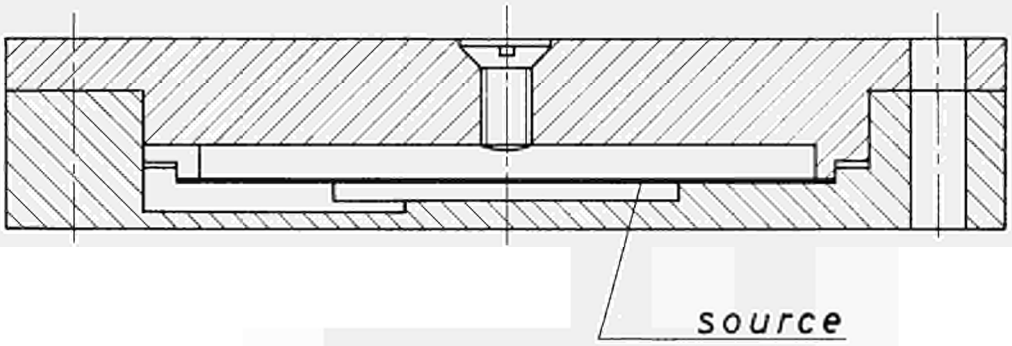

Fig. 1 — Containers for solid sources

All sources were furthermore covered uniformly on both sides with 10-20 $\mu g/cm^2$ thick evaporated gold layers, the surface conductivity of which varied from a few ohms to some kilo-ohms per cm^2.

Every source was packed in an individual perspex container (Fig. 1). These containers keep the source in a fixed position but allow airpressure equalization between the spaces below and above the source. The four containers with the sources for one laboratory are bolted together and hung on springs in a frame (Fig. 2), which is packed in a can lined with foam rubber. With

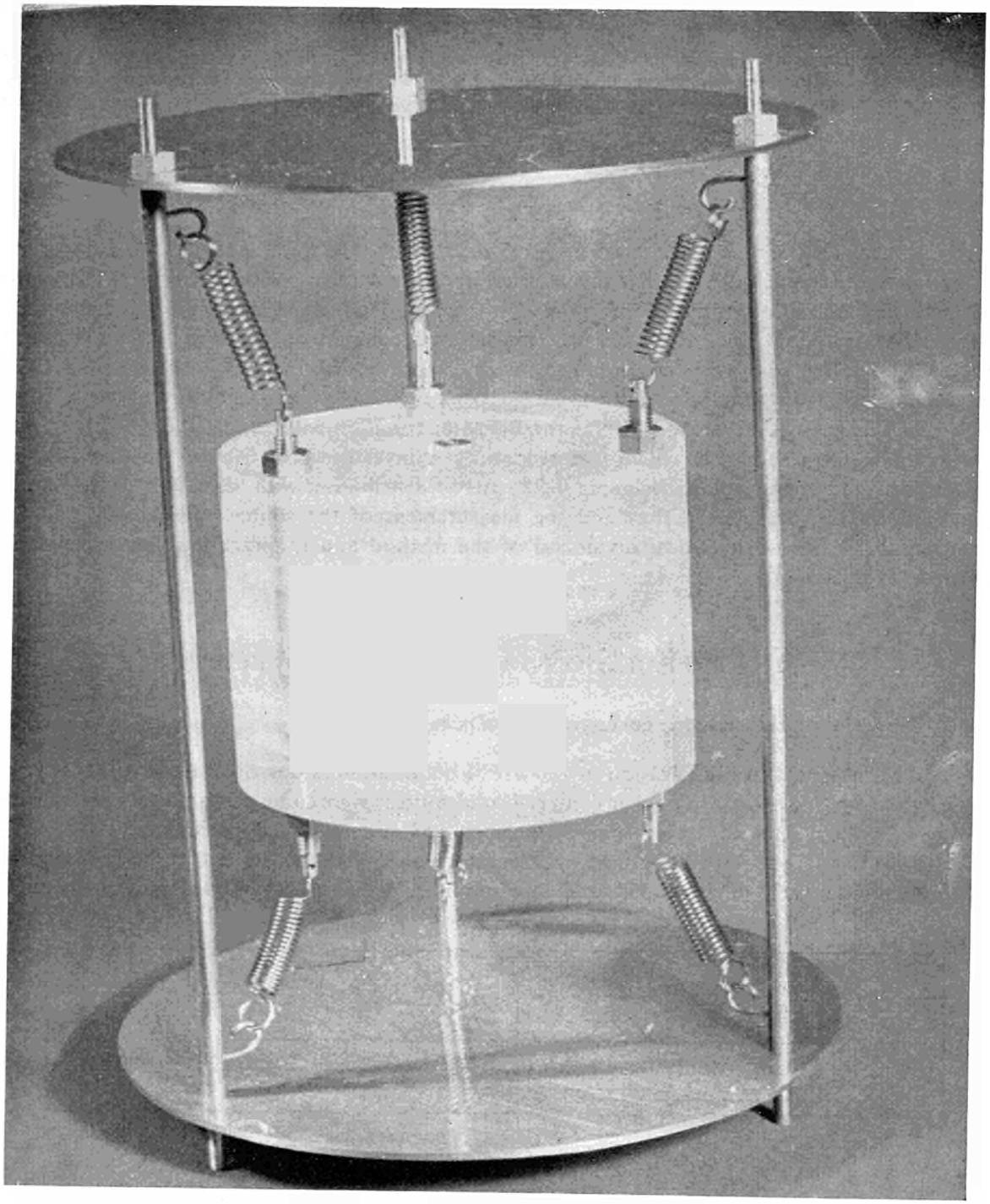

Fig. 2 — Mounting of 1 set of 4 containers

this packing, which was improved considerably during the first test comparisons, at least 90% of all sources withstood severe shocks and survived long air journeys undamaged.

2.2 — Preparation of high β-efficiency sources

The high β-efficiency sources were prepared by electrolysis on thin foils, because this is the only means of producing sources with β-efficiencies approaching 100% and extremely high mechanical stability [2-4]. The cell used for the electrolytic deposition is shown in Fig. 3. The foil employed is usually 5-10 $\mu g/cm^2$ thick VYNS of 2×2 cm surface layed down on the standard aluminium mounts. The surface attached to the mount was coated with a gold layer before and the upper side after deposition on the mount. After the foil is placed in the cell, a 20-40 mg drop of the cobalt solution used is applied to the foil and a drop of 10% NH_4OH solution is immediately added in order to keep it at a P_H of 8 or slightly over. A capillary with an inside

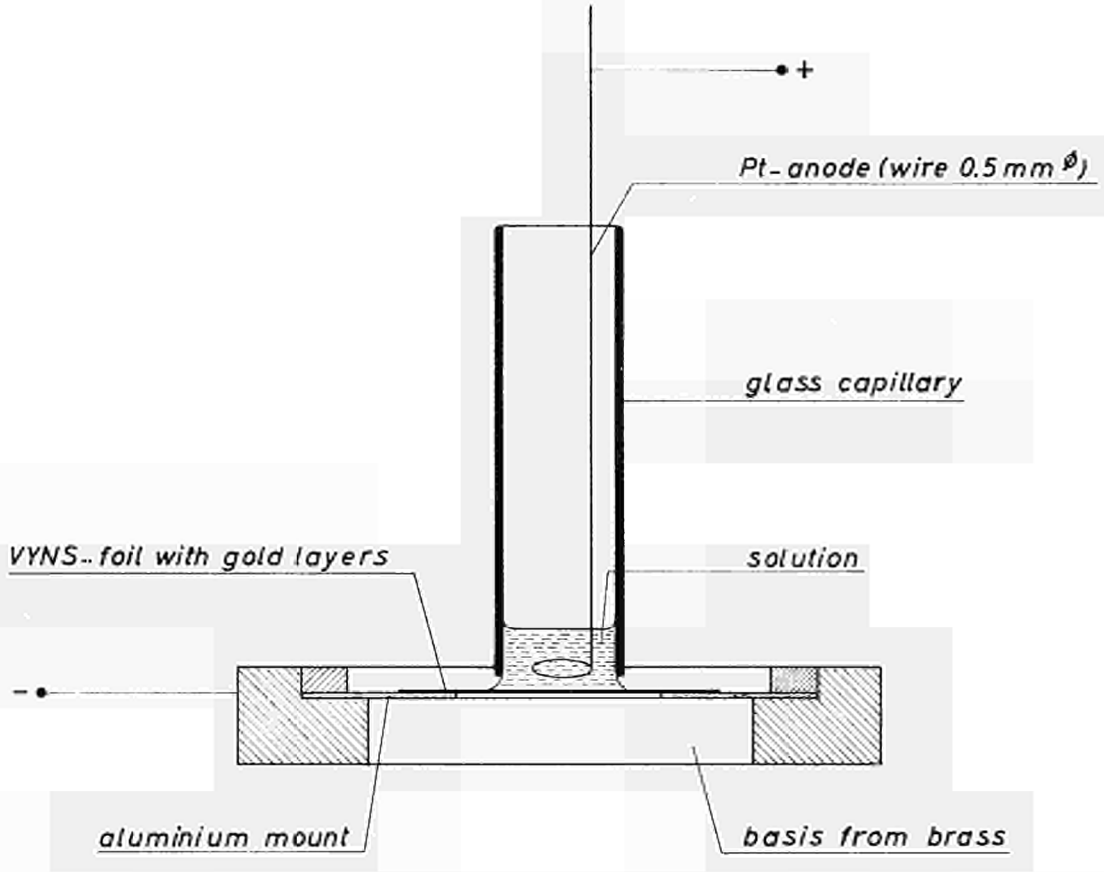

Fig. 3 — The electrolytic cell used

diameter of 6-8 mm is then placed above the drop and lowered till it contains the drop completely. A platinum-ring anode is thereupon placed about 2 mm above the foil and a current of approximately 80 mA is allowed to flow for 5-10 minutes at 8 Volts. If necessary, some NH_4OH is added with a pipette from the side in order to keep the P_H slightly above 8. After electrolysis, the foil is rinsed with distilled water, whereupon a large drop of distilled water is deposited on the foil and left for a half hour, after which the foil is thoroughly rinsed again with water. The efficiency of this method of electrolytic deposition was 30-70% and we did not succeed in increasing it to 100%. Consequently these sources cannot be used for determining the activity of the solution, as would be desirable in comparisons of this kind.

The Co^{60} solution used was a CKS 1 sample from Amersham of May 1960, diluted with 0.1 N HCl to approximately 10 μC/ml and 5 μg Co/ml. The β-efficiency of the sources was approximately 98%, which value is determined by the thickness of the foil and gold layer used. These were chosen as small as was compatible with the desired mechanical strength. 300 sources were prepared for the BIPM distribution in a period of 1-2 weeks.

2.3 — Preparation of 50% β-efficiency sources

The 50% β-efficiency sources were prepared from a well-known solution by simple drop-deposition and precipitation. A 3 mg/cm²-thick foil of acetylbutyrate or "Makrolon" was coated with a gold layer and placed with the coated side against a standard aluminium mount. A weighed drop of the Co^{60} solution — the same as was used for the electrolysis — was then pipetted onto the bare foil side, whereupon a drop of 10^{-4} Ludox solution followed by a drop of 20 μC/ml $K_3Fe(CN)_6$ solution were added. The crystals of the sources were fixed in position by covering the source with a drop of a solution of 1% cellulose in amyl acetate and drying. A foil on an aluminium mount identical with that used for the source preparation was finally used, with the gold layer outside, for covering the prepared source. The two aluminium rings were spot-welded at eight points for mechanical stability and electrical contact. The β-efficiency of these sources was approximately 48%.

3 — PROPERTIES OF THE SOLID SOURCES

3.1 — Mechanical stability, grain size, weight

The electrolytically deposited cobalt layers adhere firmly to the gold layer. No structure is recognizable under the microscope, even with a magnification of × 1000. The weight of the foils increases during electrolysis by several micrograms, but approximately the same increase occurs if a drop of distilled water is applied to the foil for a period equal to the duration of the electrolysis. (It was proved that the VYNS foils absorb water up to 30% of their weight). From these results the weight of the cobalt deposit can be estimated at less than 1 μg. The weight of the VYNS foils and of the gold layers was checked from time to time by weighing, so that the figures given in the calibration sheets can be taken as accurate to within ± 3 μg/cm².

3.2 — Constancy of the source activity

For a comparison of solid sources it is very important that their activity should remain constant over a long period and be independent of their treatment. Several series of measurements were therefore performed in order to prove that our sources meet these requirements to a high degree. 30 test sources were first measured in August 1962 and again two months later. As table 1 shows, the difference between the means of the two activity measurements was 0.004% and the mean difference between the two results for one source was 0.13%, the statistical accuracy in both measurements being within 0.1%. Similar results were obtained for 9 of these sources after 10 months (table 1). Two of these sources were mechanically shaken for several hours under different conditions and subjected to severe shocks, but their activity remained constant within 0.1%, i.e. the statistical error. Finally, 18 sources were measured with a statistical accuracy of within 0.05% before and after air transport to England. The two series of measurements (table 2) gave a mean difference of 0.06% for the single source. From all these results it can be concluded with absolute certainty that the activity of the sources used does not vary over many months

by more than 0.1%, even under extreme conditions such as severe shocks, air transport, several measurements at high voltage, etc.

TABLE 1

Results of the initial measurements of 30 sources in August 1962 and of the subsequent measurements 2 months and 10 months later

No.	Source No.	Activity (μC)			Activity ratio	
		Aug. 62	Oct. 62	June 63	Aug./Oct.	Aug./June
1	3994	0.13255	0.13234		1.00159	
2	3996	0.08783	0.08756		1.00308	
3	4010	0.07267	0.07276		0.99876	
4	4011	0.09919	0.09935		0.99839	
5	4012	0.10647	0.10666	0.10648	0.99822	0.99991
6	4013	0.06998	0.06999	0.06990	0.99986	1.00114
7	4014	0.09647	0.09670	0.09656	0.99762	0.99907
8	4015	0.08536	0.08541		0.99941	
9	4018	0.06133	0.06133	0.06144	1.00000	0.99821
10	4033	0.10632	0.10640	0.10628	0.99925	1.00038
11	4034	0.07475	0.07472		1.00040	
12	4035	0.11094	0.11099		0.99955	
13	4036	0.09325	0.09303		1.00236	
14	4039	0.11511	0.11502		1.00078	
15	4040	0.10067	0.10096		0.99713	
16	4041	0.13455	0.13471		0.99881	
17	4056	0.09839	0.09822		1.00173	
18	4065	0.13704	0.13669	0.13698	1.00256	1.00044
19	4066	0.10652	0.10629		1.00216	
20	4068	0.08337	0.08338		0.99988	
21	4069	0.07104	0.07080		1.00339	
22	4073	0.07413	0.07403		1.00135	
23	4074	0.11047	0.11051		0.99964	
24	4075	0.12369	0.12375		0.99952	
25	4076	0.12816	0.12818		0.99984	
26	4080	0.05967	0.05973		0.99900	
27	4081	0.09050	0.09060		0.99890	
28	4082	0.13223	0.13222	0.13224	1.00008	0.99992
29	4084	0.13726	0.13718	0.13711	1.00058	1.00109
30	4085	0.13579	0.13613	0.13588	0.99750	0.99934

3.3 — The β-efficiency, its constancy and absolute value

The activity is a characteristic property of the source alone and should therefore be reproduced in all measurements of different kinds and under different conditions. This does not apply to the β-efficiency, which in general is defined and measured for a certain overall discrimination of the counting apparatus. However the detection of β-particles depends very largely on this discrimination level. Consequently β-efficiencies cannot be compared as easily as activities. Nevertheless, it is important to know the constancy of the β-efficiency if it is measured under the same conditions. We therefore measured the β-efficiencies of 18 sources (table 2) before and after air transport to England, using the same counting equipment. The two results agree within the statistical error of 0.05%. As in the case of the activities the conclusion is that our sources are also stable to within at least 0.1% as regards their β-efficiency.

The absolute value of the β-efficiencies of our electrolytically deposited sources at approximately 1 keV discrimination is 97.2 to 98.0%. This value can be explained satisfactorily. Firstly

TABLE 2

Results of measurements before (March) and after (April) air-transport

No.	Source No.	Activity (dps)			β-efficiency (%)		
		March	April	March/April	March	April	March/April
1	5083	3521.2	3520.3	1.00026	97.62	97.54	1.00082
2	5114	1974.4	1973.2	1.00061	97.69	97.80	0.99888
3	5160	2525.1	2524.7	1.00016	97.82	97.72	1.00102
4	5161	3506.4	3510.0	0.99897	98.05	97.88	1.00174
5	51721	2606.2	2603.6	1.00100	97.94	97.95	0.99990
6	5196	2671.9	2671.0	1.00034	97.83	97.82	1.00010
7	5201	3605.7	3605.5	1.00006	97.63	97.70	0.99928
8	5202	4053.9	4053.4	1.00012	97.68	97.68	1.00000
9	5211	4239.3	4239.3	1.00000	97.62	97.65	0.99969
10	5222	1325.4	1326.3	0.99932	97.90	98.02	0.99878
11	5223	1461.4	1461.2	1.00014	97.77	97.80	0.99969
12	5270	3493.2	3493.7	0.99986	98.07	98.12	0.99949
13	6573	1176.5	1179.7	0.99729	97.85	98.03	0.99816
14	6588	947.8	950.5	0.99716	97.75	97.79	0.99959
15	51751	6056.7	6055.5	1.00020	48.51	48.53	0.99959
16	51881	5543.0	5544.2	0.99978	48.33	48.35	0.99959
17	51891	5283.0	5281.8	1.00023	48.53	48.51	1.00041
18	6809	4906.7	4910.8	0.99917	48.87	49.06	0.99613

TABLE 3

Reproducibility of the γ-efficiency in a series of measurements on 12.3 - 23.3.1963 (statistical error 0.1%)

No.	Source No.	ε_γ(%)	$\varepsilon_{\gamma/\bar{\varepsilon}\gamma}$	No.	Source No.	ε_γ(%)	$\varepsilon_{\gamma/\bar{\varepsilon}\gamma}$	No.	Source No.	Source	$\varepsilon_{\gamma/\bar{\varepsilon}\gamma}$
1	5039	33.030	0.99997	11	5109	33.033	1.00006	21	5186	33.018	0.99961
2	5040	33.056	1.00076	12	5065	33.051	1.00061	22	5190	33.054	1.00070
3	5055	32.991	0.99879	13	5122	33.057	1.00079	23	5194	33.041	1.00030
4	5083	33.023	0.99976	14	5125	33.052	1.00064	24	5195	33.047	1.00048

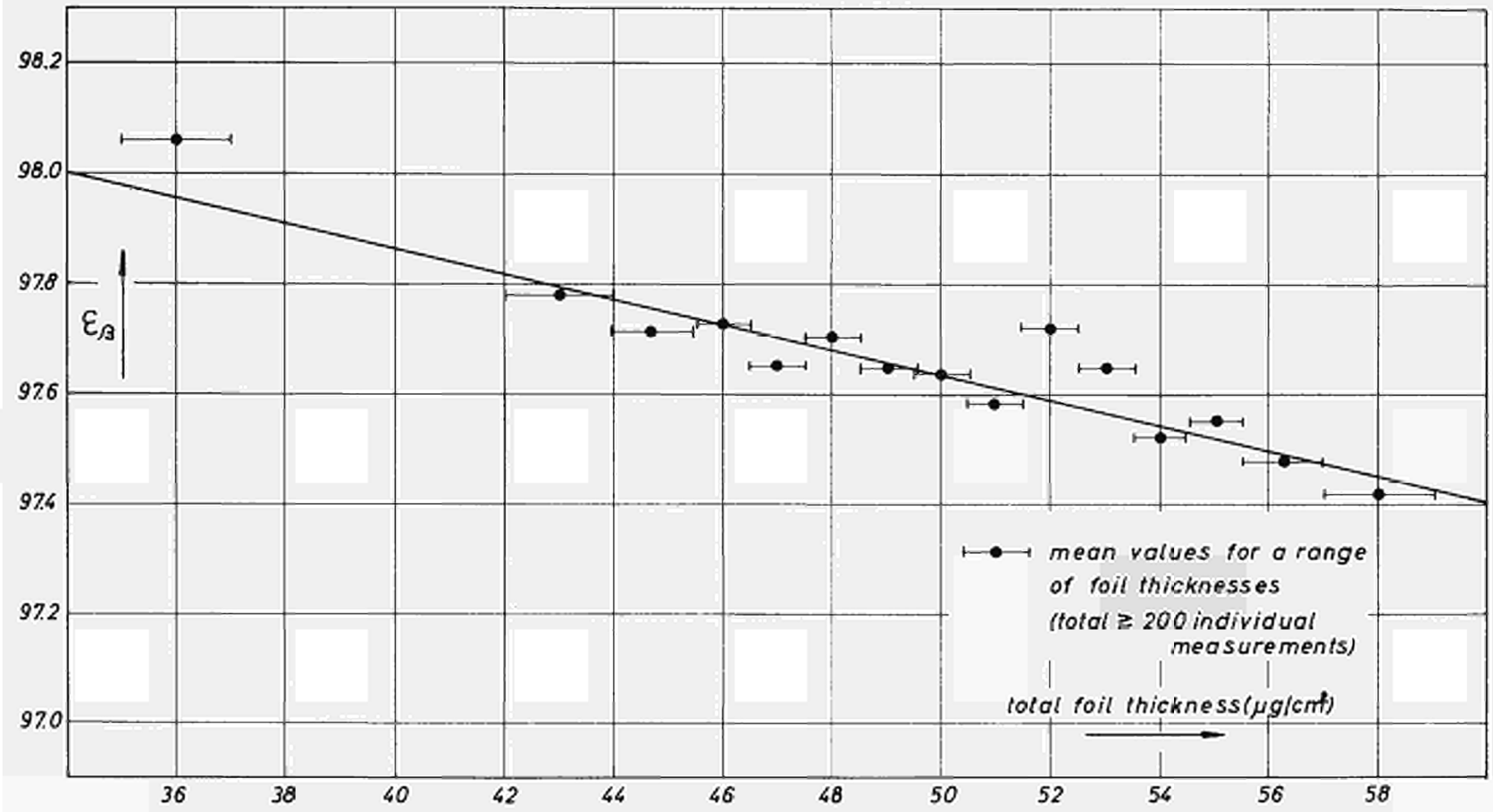

Fig. 4 — Dependence of the β-efficiency on the total foil thickness

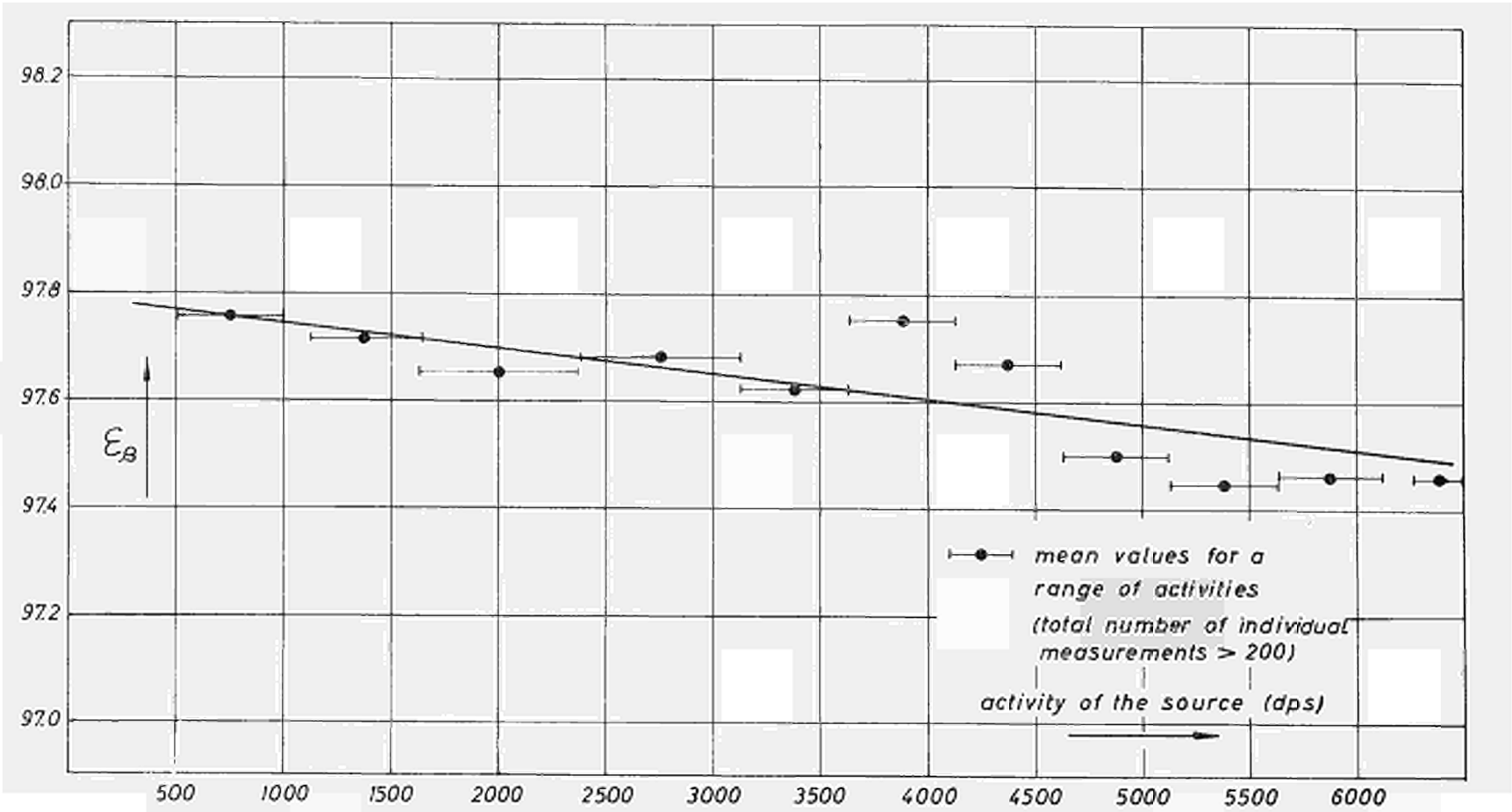

Fig. 5 — Dependence of the β-efficiency on the activity of the source

3.4 — The γ-efficiency

The γ-efficiency is a characteristic property of the counting apparatus alone (at least for a thin point source) and is therefore not intercomparable. As an apparatus constant it is subjected only to source-independent statistical fluctuations, as is confirmed by the experimental results in table 3.

4 — PREPARATION AND PROPERTIES OF THE DISTRIBUTED SOLUTION

Two CKS 1 solutions from Amersham dating from May 1960 and November 1962 were mixed and diluted with 0.1 N HCl containing 30 μg/ml $CoCl_2$ to an activity of approximately 1,500,000 dps/ml and a carrier content of approximately 20 μg/ml Co; they were then used for the distribution. A γ-spectrometer test revealed less than 0.05% γ-impurities with energies above 30 keV; a half-life check on the older solution over a 3 years period gave a half-life of 5.265 ± 0.005 years. The radiochemical purity can thus be guaranteed to within 0.1%. The activity of the solution as measured respectively by the $4\pi\beta$-γ-method and the liquid scintillation method was $1525.0 \pm 0.2\%$ and $1526.8 \pm 0.2\%$ dps/mg on 1.1.1963. A check on these results with ionisation chamber having an accuracy of within 2% gave 1534 dps/mg. The solution was distributed in 5-ml ampoules. The latter were weighed empty and again after filling (by means of an automatic burette) and sealing. This method of filling and weighing was accurate to within

TABLE 4

Comparison between weight and γ-activity of solution in the ampoules

No. amp.	Sent to	mg	$\frac{\text{cps/mg}}{\overline{\text{cps/mg}}}$	No. amp.	Sent to	mg	$\frac{\text{cps/mg}}{\overline{\text{cps/mg}}}$
1	IAEA	4350.4	1.00066	28	EL	4831.2	0 99820
2	IAEA	4841.1	0 99896	29	NPL	4878.2	1.00047
3	JEN	5017.7	0.99924	30	NPL	4783.8	0.99820
4	JEN	4801.0	0.99953	31	AEET	4917.3	0.99981
5	GWI	4739.8	1.00114	32	AEET	4835.3	0.99848
6	GWI	5073.4	0.99962	33	DAECRE	4766.7	1.00152
7	IRK	4962.8	0.99905	34	DAECRE	5009.5	0.99659
8	IRK	4963.2	0.99962	35	BIPM	4699.7	0.99924
9	IPA	4933 6	1.00152	36	BIPM	4628.9	1.00057
10	IPA	4864.4	1.00142	37	IMM	4746.7	0.99867
11	IFM	4829.3	1.00057	38	IMM	4696.5	1.00398
12	IFM	4834.7	0.99858	39	NBS	4718.9	0 99829
13	CBNM	3854.8	1.00341	40	NBS	4696.8	1 00000
14	CENS	4837.4	0.99886	41	LNE	4690.6	1.00142
15	NPRL	4798.4	1.00038	42	LNE	4656.8	0.99972
16	CENS	4861.5	0.99915	43	AECL	4832.6	1.00322
17	ISS	4803.0	1 00208	44	NPRL	4747.0	0 99934
18	ISS	5051.1	0 99934	45	AECL	4716.7	1 00436
19	UVVVR	4911.1	1.00133	46	IKO	4764.3	1.00161
20	UVVVR	4919.1	0 99886	47	PTB	4767.5	0.99886
21	AAEC	4822.6	1.00076	48	PTB	4805.2	0.99706
22	AAEC	4918.9	0 99801	49	IKO	4918.8	0.99991
23	IAR	4951.6	0 99877	50	CBNM	4569.4	1.00000
24	IAR	4805.3	1.00047	51	ISN	4472.7	—
25	NRC	4779.4	1.00009	52	ISN	4669.8	—
26	NRC	4801.2	1.00009	53	not	4559.7	—
27	EL	4844.1	0.99801		used		

0.02%, as was proved by a series of inactive tests. A further check on the amount of solution in the ampoules was obtained by measurement of all the ampoules in a γ-spectrometer which was accurate to within 0.05%. Table 4 shows the results, all of which agree to within 0.35% with the results of the weighing. This is the normal mean difference for different ampoules. We can therefore guarantee that the figures quoted for the activity and the quantity of solution in the ampoules are correct to within approximately 0.1%.

5 — APPARATUS AND TECHNIQUE FOR THE $4\pi\beta$-γ-MEASUREMENT

5.1 — Counter

The counter used is shown in Fig. 6. It was constructed after several years' experience with other counters and has the following principal characteristics : γ-efficiency for Co^{60}, 33-34% at 20 keV discrimination; reproducibility of the γ-efficiency, 0.1%; crystal diameter, 3″; crystal height, 2″ (chosen for an optimum ratio between γ-efficiency and γ-background); lead shield

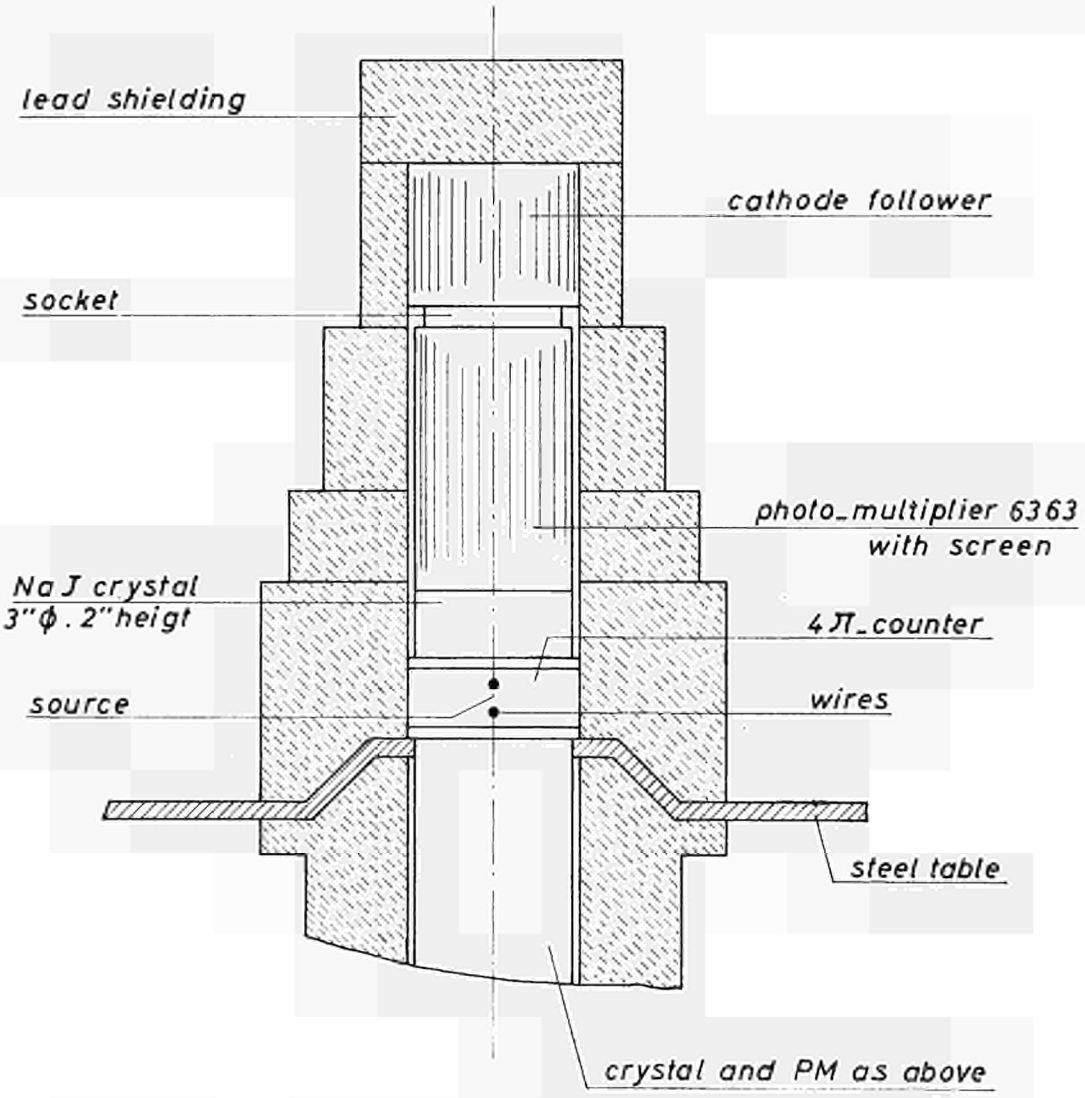

Fig. 6 — Final $4\pi\beta$-γ-counter used (upper part; the bottom part is symmetrically the same)

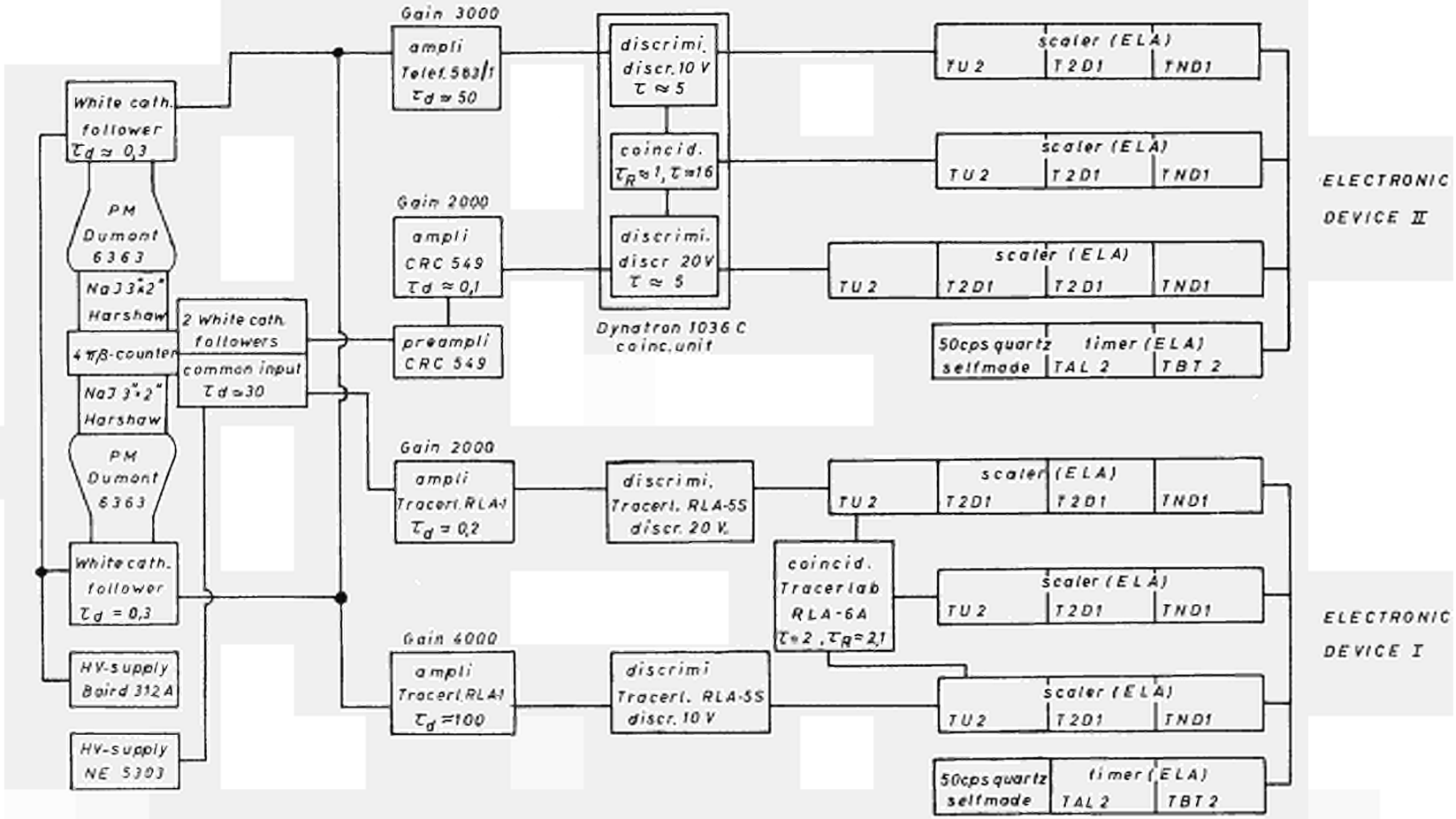

Fig. 7 — The two counting chains used in parallel (τ_d differentiation time constant, τ dead time, τ_R coincidence resolution, all in μs)

surrounding the crystal, 6 cm (chosen from the measured dependence of the γ-background on the shield thickness where at 6 cm the decrease of the background with increasing lead thickness begins to get low).

5.2 — Counting electronics

Previous experience showed that our electronic counting equipment gives considerable additional trouble when the counting accuracy is increased from within 1% to within 0.05%. We therefore always work with two complete counting chains (denoted as I and II) in parallel (Fig. 7). This method has proved very useful [5], since it usually enables defects to be traced very quickly. Furthermore, it can be used for checking the most important corrections, provided that the experimental conditions can be so chosen that these corrections are independently and differently determined in the two electronic devices. This advantage is applied in our equipment for the determination of the dead-time correction (see 7.4).

The choice of the characteristic constants of the electronic counting apparatus is closely connected with the main problem of high-precision coincidence counting, namely the avoidance of any spurious pulses or loss of true pulses in the individual counting channels. The amplification in the β-channel should be as high as possible in order to meet the requirements of a $4\pi\beta$-method (4π actually means that the β-efficiency is 100%). Care must, however, be taken that the amplifier does not produce afterpulses. This requirement is met only by low-noise amplifiers with extremely good overload performance, mainly non-linear or transistor types. The same considerations apply to the choice of the various time constants of the counting electronics, namely differentiation time constant, integration time constant, dead times in the 3 channels and the resolving time of the coincidence. The differentiation time constant must be 2-3 times as high as the rise time of the detector, in order to prevent the pulse height from varying according to its point of origin [6]. The integration time constant must be of the magnitude of the differentiation time constant for a good signal-to-noise ratio [6]. The dead times should be as short as possible, but care must be taken that every afterpulse or overshot falls into the corresponding dead time. Also the resolving time of the coincidence should be as short as possible (to reduce the correction for accidental coincidence) but it must be absolutely certain that no coincidences are lost. Since the choice of these time constants and their positioning in the counting chains are rather difficult problems, the results should always be tested experimentally. We did this by measuring the decay rate N_0 as a function of the high voltage in the β-channel (control of the differentiation and integration and the resulting overload characteristics), as a function of the dead times and as a function of the resolving time of the coincidence (Fig. 8, 9 and 10). The constants finally chosen are given in Fig. 7. There is still one electronic adjustment to be discussed, namely the choice of the discrimination in the γ-channel. Two methods of γ-discrimination appear particularly advantageous : integral discrimination at a value so low that this is mainly determined by the absorption in the window of the photomultiplier, and differential discrimination over one or more of the photo-peaks of the γ-radiation [7-10].

Since the first method is simple and does not depend on fluctuations of the discrimination level and the second is only of special value in measurements of low activities or complicated decay schemes, we chose the first method for the Co^{60} measurements. The effective discrimination level was then set at 25 keV and checked with γ-rays from Am^{241}.

Some general precautions were found necessary in 0.1% precision counting, in particular the use of a quartz timer, stabilisation and filtering of the line voltage supply, and checking of the timer. The latter was carried out by feeding standard quartz-oscillator pulses to a counter which was started and stopped by the timer used. This method revealed, for example, that the stop pulse needed 0.1 sec before triggering, so that even in measurements of 100 sec duration a correction of 0.1% must be applied to the counting time.

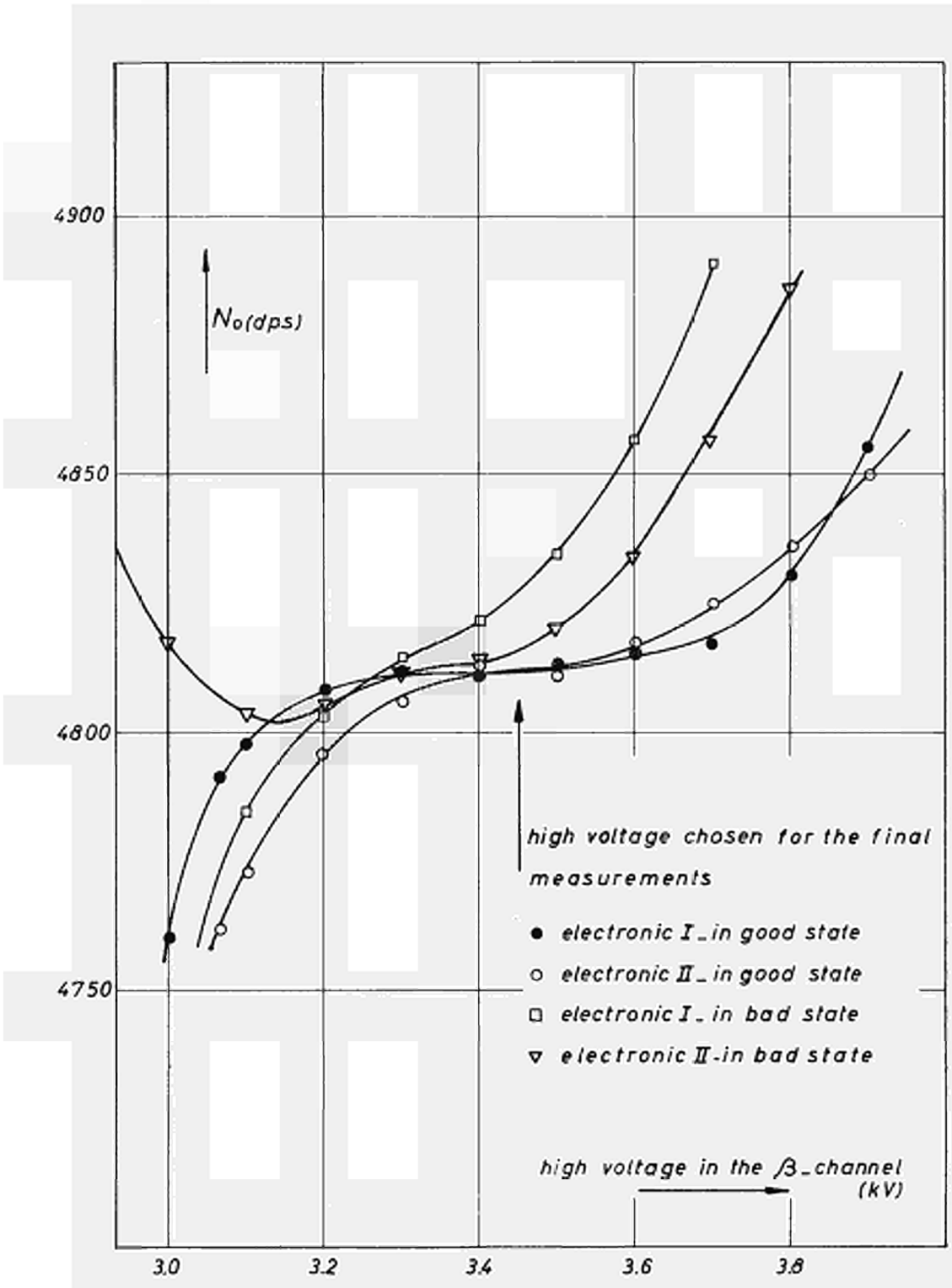

Fig. 8 — Examples of decay-rate dependence on the high voltage in the β-channel (N_o-plateaux)

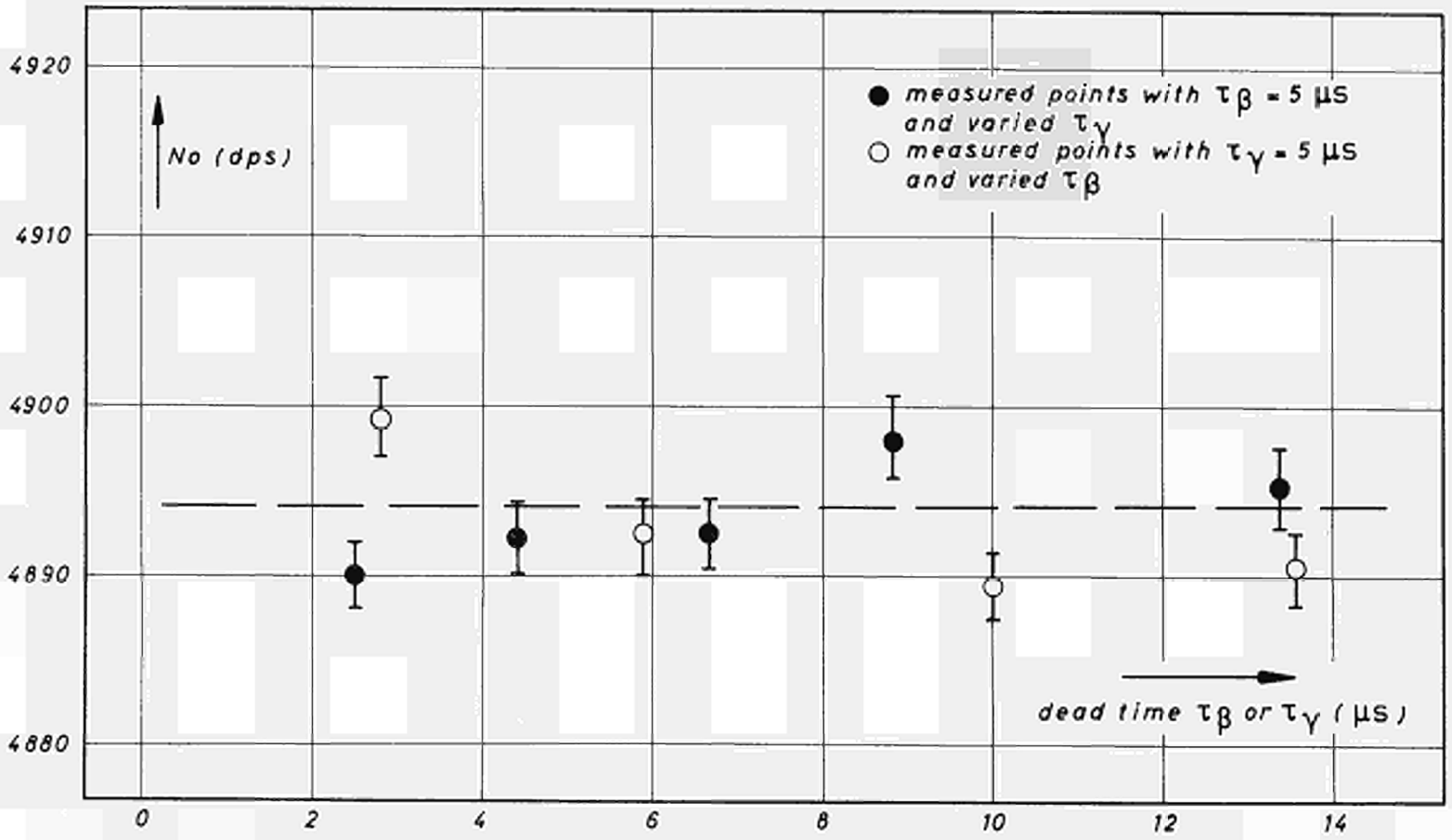

Fig. 9 — Control of dead time corrections (electronic device I; similar results for II)

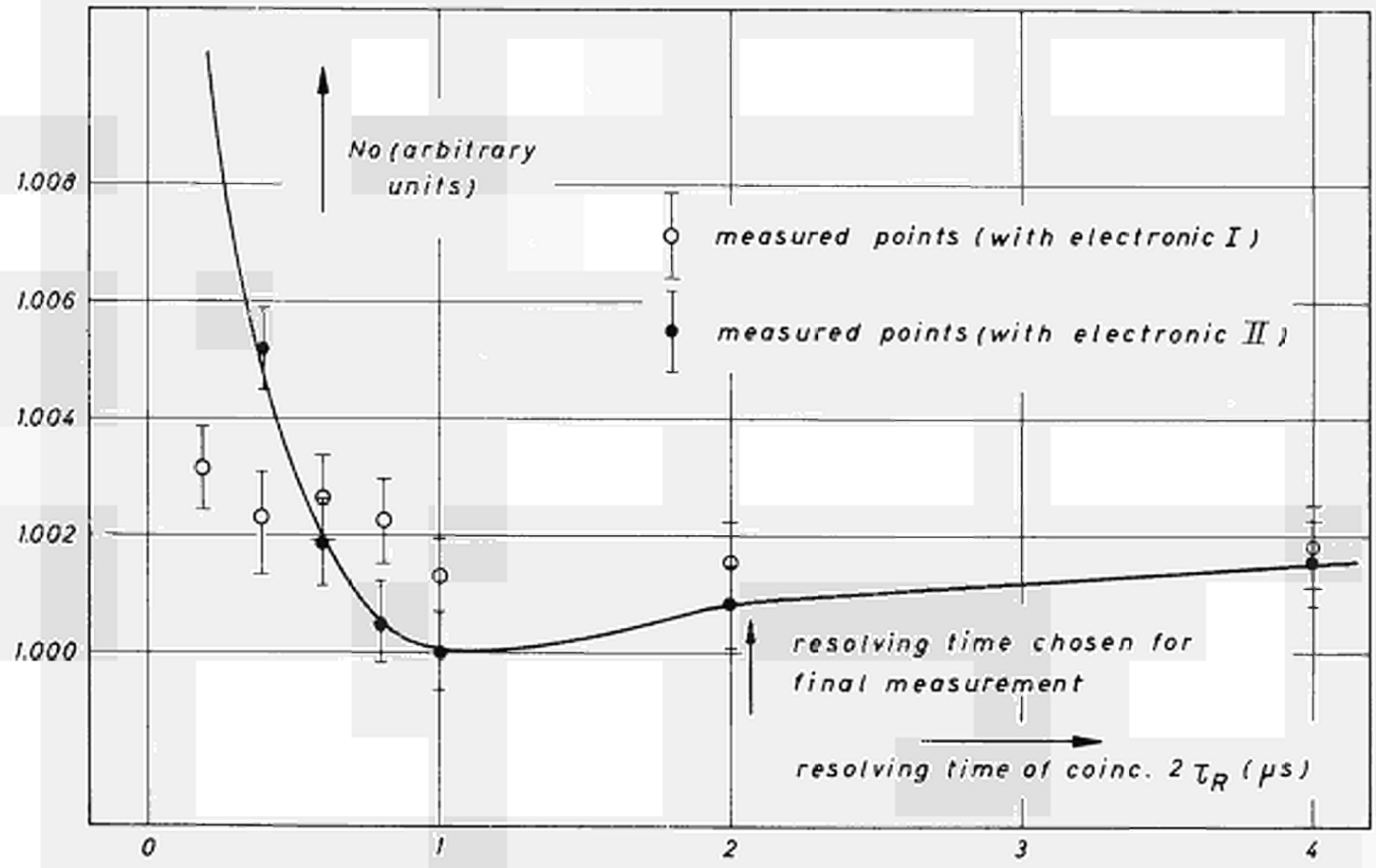

Fig. 10 — Measured decay rate as a function of the resolving time of the coincidence

5.3 — Daily testing and reproducibility of the counting apparatus

Since previous extensive measurements had shown clearly that a precision of within 0.1% can only be guaranteed if the counting apparatus is tested frequently, all constants which enter into the calculations of the activity were measured at least once, and for the most part twice, daily. These tests started with a check on the amplifications by feeding a standard pulse from a Model 370 Franklin pulse generator into the test input at the output of the photo-multiplier. The dead times in the 3 channels were then measured with a Beckmann Model 4094 double pulse generator and an oscillograph, and the resolving time of the coincidence unit was determined with two independent pulse generators for the β- and γ-channels and by calculation with the formula $2\tau_r = N_c/N_\beta N_\gamma$. Thereafter normal background measurements were performed, each time followed by a 10×100 sec counting of a standard cobalt source of approx. 5000 dps. The results of these tests not only served for checking the stability of the counting equipment, but were also collected for a determination of the fluctuations of the constants, these being used as the basis for the statement of the measurement errors. For these fluctuations we obtain the following results (measurement period December 1962 - February 1963) : the standard deviations of the amplifications were a few per cent (no appreciable effect on the discriminations); the standard (mean) deviation of the single measurement of the β-background was 10% and that of the γ-background 4%; the standard deviations of the dead times were 10% in β-channel I, 15% in γ-channel I, 5% in coincidence channel I, 1.6% in β-channel II, 9% in γ-channel II, 1.3% for coincidence channel II; the stand. deviations (of single measurements) of the resolving time were 6% for channel I and 0.5% for channel II.

The fluctuation in the activity of the standard source is very interesting because this characterizes the precision of the measurement. Here the results obtained were not as good as we had hoped in view of all the precautions taken. Although the standard deviation of the activity results for a single source was always below 0.1%, we got fairly high fluctuations : for example, a standard deviation of 0.04% with a statistical accuracy of within 0.05%; a standard deviation of 0.08% with a statistical accuracy of within 0.03%; and a standard deviation of 0.1% with a statistical accuracy of within 0.05% (at least 10 individual measurements in each case). The conclusion is that we must increase our efforts if we desire to achieve a counting-reproducibility of below 0.1%, and that this is the limit for our present measurements.

6 — FORMULAE FOR EVALUATING THE RESULTS OF A $4\pi\beta$-γ-COUNT

6.1 — Formula for the decay rate

Most of the fundamental formulae for $4\pi\beta$-γ-measurements are derived and proved in Campion's basic paper [1]. In the present document, therefore, we shall merely mention these basic facts (and the corresponding definitions) and discuss in detail only the checks, extensions and possible minor improvements.

The decay rate N_0 is determined under ideal conditions of geometry, decay scheme, etc., from the known formula $N_0 = N_\beta N\gamma/N_c$. The corrections necessary in practice can be divided into backgrounds, accidental coincidences, count-rate-dependent corrections (= dead-time corrections) and count-rate independent corrections (= decay-scheme-dependent corrections). If these corrections are applied wherever possible as factors (for easier machine calculations) we get the following formula for the decay rate :

$$N_0 = N_\beta N_\gamma K_\varepsilon K_d/(N_c - N_{\mathrm{acc}}) \tag{1}$$

where N_i are the measured rates corrected for backgrounds ($N_i = N_i' - B_i$), N_i' are the directly

measured rates in the channels i and B_i the corresponding backgrounds, N_{acc} is the accidental coincidence rate and K_ε and K_d are the corrections for the decay-scheme-dependent effects and for dead-time effects respectively. These corrections are discussed in detail in the following sections.

6.2 — Formulae for the efficiencies

Various authors have given widely differing definitions of the β- and γ-efficiency of a counting device. The only definition that characterizes the detector unequivocally is : the number of radiations of specific kind and energy which is detected per number of radiations of the same kind created in the source. This definition normally takes account of the influence of the discrimination outside the detector, which as a rule is difficult to determine separately. For the sake of completeness therefore, this overall discrimination in the counting chain should always be stated. In accordance with the above definition we obtain the following equation for the β-efficiency :

$$\varepsilon_\beta = \{N_\beta - (N_{i\neq\beta})_\beta\}/\{N_0(1 - t_\beta N'_\beta)\}. \tag{2}$$

In the case of cobalt 60 this reduces to :

$$\varepsilon_\beta = \{N_\beta - (\varepsilon_\gamma)_\beta p_\beta N_0(1 - \varepsilon_\beta)\}/\{N_0(1 - t_\beta N'_\beta)\} = \tag{2a}$$

$$= \{N_\beta - (\varepsilon_\gamma)_\beta p_\beta N_0\}/\{N_0(1 - t_\beta N'_\beta - (\varepsilon_\gamma)_\beta p_\beta)\}. \tag{2b}$$

Here $(N_{i\neq\beta})_\beta$ is the rate of pulses counted in the β-counter but not caused by β-particles from the source. In the case of cobalt-60 these pulses can be caused only by γ-rays, so that they can be described by $(N_{i\neq\beta})_\beta = (\varepsilon_\gamma)_\beta p_\beta N_0(1 - \varepsilon_\beta)$ with an efficiency $(\varepsilon_\gamma)_\beta$ for the detection of γ-rays in the β-counter and a dead-time correction factor p_β [1] for the latter. Since $(\varepsilon_\gamma)_\beta$ is of the order of 1-2% (we measured 1.8% at 1 keV discrimination), we can neglect the terms containing it if the β-efficiency is higher than approximately 90%. For this case we get, with sufficient accuracy :

$$\varepsilon_\beta = N_\beta/N_0(1 - t_\beta N'_\beta). \tag{2c}$$

The same equation holds good for the γ-efficiency if no β-particles can be detected in the γ-detector, as is certainly the case here. We thus obtain :

$$\varepsilon_\gamma = N_\gamma/N_0(1 - t_\gamma N'_\gamma). \tag{3}$$

7 — CORRECTIONS IN $4\pi\beta$-γ-MEASUREMENTS AND THEIR VERIFICATION

7.1 — General remarks

There are two methods of determining the necessary corrections for measured results : a theoretical investigation, which leads to general correction formulae (with constants which in general have still to be determined experimentally), and a purely experimental determination, which normally results in an extrapolation method [9-12]. Since the assumptions of the theoretical methods are never completely fulfilled in practice, it would appear desirable always to employ both methods in high-precision work. This is consistently done in the present paper. In all discussions of the corrections the discrimination level in the electronic apparatus is assumed to be zero. This can generally lead to considerable errors (for example in the dead-time corrections), but it was proved in every individual case that this assumption can be made here.

Finally, it should be mentioned that the results of the correction determinations should also be used for the choice of the various constants (dead times, etc.), in order to keep the corrections as small as possible without introducing other errors.

7.2 — Backgrounds

The backgrounds are easily measured by using an empty foil and are readily corrected by subtraction. Sometimes (mainly in low-energy measurements) it must be ascertained whether high-energy background pulses cause any serious disturbances.

7.3 — Accidental coincidences

Before any discussion of the correction for accidental coincidences it must be stated exactly which pulse combinations are to be understood by that term. The definitions in the literature vary considerably. Some authors [13-16] even treat the accidentals together with the dead times, in which case the dead time in the coincidence channel is reduced by a number of accidentals which appear at the beginning and the end of dead times in the β- and γ-channels.

However separate treatment of the accidentals seems to be clearer. In this case the accidental coincidences can be defined according to Campion [1] as : the detection of a particle in each of the two counters within the resolving time t_r, whereby each particle comes from another decay and the respective second particles of the two decays are not counted owing to the limited efficiency of the counting apparatus and to dead times. (Brinkman, for example, does not regard the last effect as responsible for accidentals). This definition covers coincidences between true and background pulses, but these accidentals can be ignored in the following considerations, as is permissible for moderate γ-efficiencies [16]. The formula for the accidentals defined above is given by Campion [1] and can be proved by different other derivations. It is :

$$N_{\mathrm{acc}} = \{2t_r N'_\beta N_\gamma - t_r(N'_c - B_c)\,(N'_\beta + N'_\gamma)\}/\{1 - t_r(N'_\beta + N'_\gamma)\} \tag{4}$$

and can be simplified for high β-efficiencies to :

$$N_{\mathrm{acc}} = t_r N'_\gamma (N'_\beta - N'_c). \tag{4a}$$

These equations are derived for $t_r < t_\beta$ and t_γ on the following asumptions : the resolving time is the same whether a β-pulse or a γ-pulse enters the coincidence unit first; all other corrections (including dead times) are ignored which is the reason why the formula contains accidentals which cannot occur (e.g. those in which one pulse of the first pair is not counted owing to a dead-time, but this continues during the second pair of pulses and thus prevents a coincidence from being counted); the background in the coincidence channel is subtracted once and then partly corrected again in the accidentals. Because of these simplifications it is useful to find other conditions of measurement which lead to the same correction in a different way. This is possible with the choice of a resolving time t_r which is higher than the dead times in the β- and γ-channels t_β and t_γ. In this case the formula for the accidentals can be derived on the same assumptions as above in a manner similar to Campion's [1] derivation, if we assume that the first-counted pulse no longer prevents the counting of the second pulse in the same channel by dead times. We then get, in Campion's [1] notation :

$$N_{\mathrm{acc}} = (N'_\gamma - N_c)t_r N'_\beta(1 - \varepsilon_\gamma p_\gamma) + (N'_\beta - N_c)t_r(1 - \varepsilon_\beta p_\beta)\,N'_\gamma = \tag{5}$$

$$= 2t_r(N'_\gamma - N_c)\,(N'_\beta - N_c). \tag{5a}$$

The dead time in the coincidence channel is of no importance in the consideration of the accidentals. It is now very interesting to check experimentally the equations derived for the accidentals. This can best be done by varying the resolving time and extrapolating to zero. Such measurements are in progress, and the initial results for a limited range of resolving times are given in Fig. 10, for $t_r < t_\beta, t_\gamma$. These show that the use of the formulae derived leads to results for the decay rate which are independent of t_r to within 0.1% at least. The formulae for the accidentals can

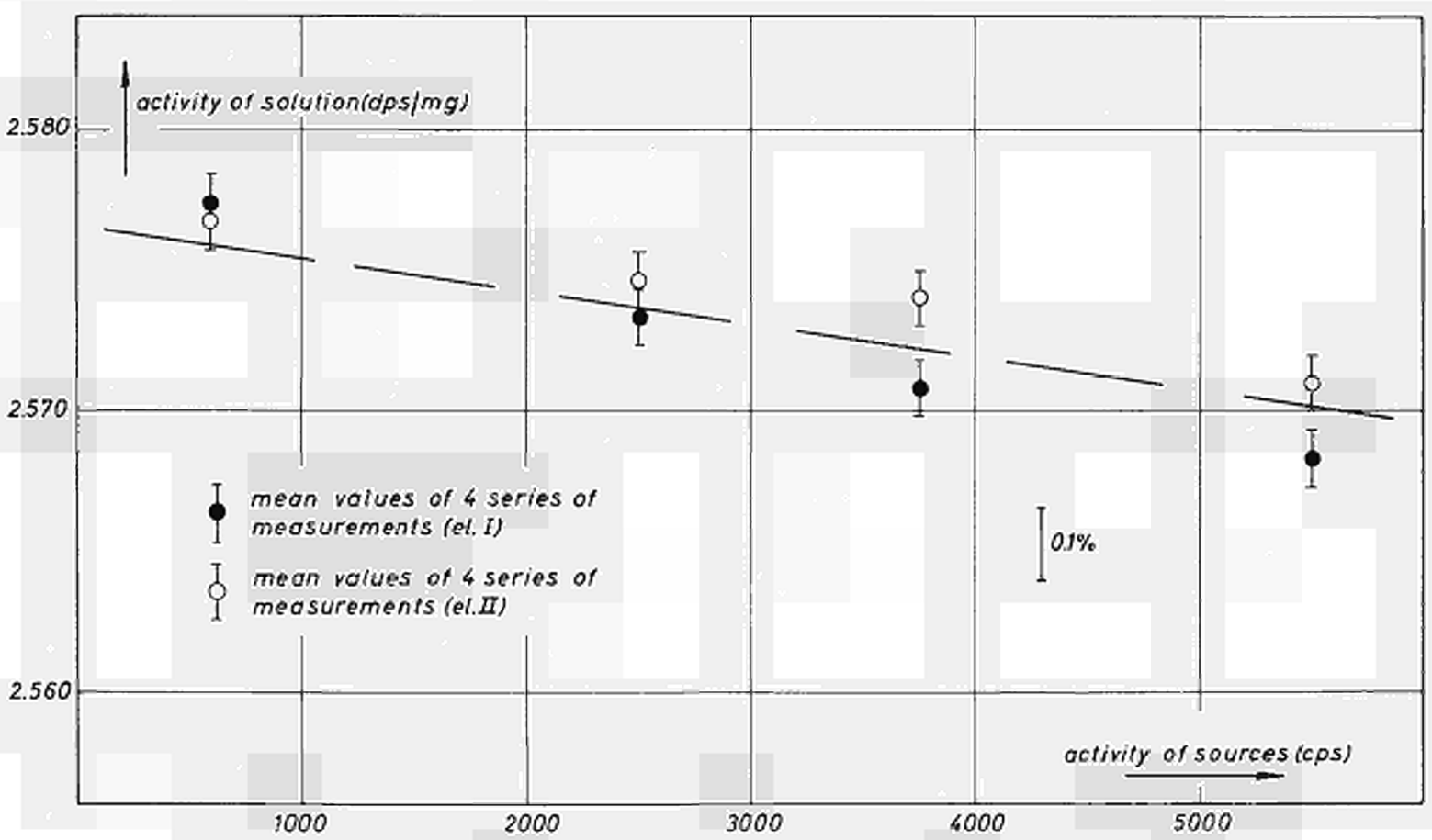

Fig. 11 — Results of decay rate measurements with sources of different activities prepared from the same solution

be further checked by measurements of short-lived nuclei [15], measurements of sources of different activities prepared from the same known solution, and measurements with different efficiencies if we assume that the dead time corrections applied are right. Fig. 11 shows the results for the activity of a solution derived from measured activities of sources of different magnitude. Since we are very sure of our dead-time corrections (see 7.4.), it can be concluded from this figure that the correction for accidentals must be also accurate to within $\pm 0.15\%$. An increase of the order of 0.1% with lower source strength must however still be explained.

7.4 — Dead times

Campion [1] has also derived a precise dead-time correction for the case $t_r < t_\beta$ and t_γ. Once again, some assumptions were made in this derivation (and must be made in most others); These are : the dead times are non-extendable (or, according to Evans [17] the counter is non-paralysing); only the first terms of the exponential functions from the Poisson distributions are taken into consideration for the calculations; in the case of unequal dead times in the β- and γ-channels, the extension of the effective dead time in the coincidence channel by coincidence-coincidence-series and coincidence — single-pulse — coincidence effects in which the second pulse falls into a period where the one channel is already open but the other still dead, is neglected. Here again, therefore, it is very useful to find other conditions for an additional determination of the dead-time correction in a different and independent way. This is again possible if t_r is chosen higher than the dead times in the β- and γ-channels. (In the case of dead-time investigations t_r always denotes the maximum of the resolving and dead times in the coincidence channel.) The dead-time correction can be derived very simply for both cases (t_r lower or higher than t_β and t_γ) in the following manner : In the single channels the total dead times are $t_\beta N'_\beta$ and $t_\gamma N'_\gamma$ respectively and the corresponding correction factors $1/(1 - t_\beta N'_\beta)$ and $1/(1 - t_\gamma N'_\gamma)$ respectively. The coincidence channel is effectively dead for $t_r < t_\beta$ and t_γ when a single channel is dead, and so the total dead time in this channel is $t_\beta(N'_\beta - N'_c) + t_\gamma(N'_\gamma - N'_c) + N'_c\, t_{\max}$, with $t_{\max}$ the maximum of the β- and γ-dead-times. With the corresponding correction we get :

$$K_d(t_r < t_\beta, t_\gamma) = \{1 - t_\beta(N'_\beta - N'_c) - t_\gamma(N'_\gamma - N'_c) - t_{\max} N'_c\}/\{(1 - t_\beta N'_\beta).(1 - t_\gamma N'_\gamma)\} \tag{6}$$

$$= 1 + t_{\min}\ N'_c + \ldots \tag{6a}$$

Here $t_{\min}$ is the lower of the two dead times t_β and t_γ. For the case where $t_r > t_\beta$ and t_γ we get the same dead times in the single channels and in the coincidence channels for the whole dead time $t_\beta(N_\beta - N'_c) + t_\gamma(N_\gamma - N'_c) + t_r N'_c$, so that for the correction we have :

$$K_d(t_r > t_\beta, t_\gamma) = \{1 - t_\beta(N'_\beta - N'_c) - t_\gamma(N'_\gamma - N'_c) - t_r N'_c\}/\{(1 - t_\beta N'_\beta)\,(1 - t_\gamma N'_\gamma\} \tag{7}$$

$$= 1 - N'_c(t_r - t_\beta - t_\gamma) + \ldots \tag{7a}$$

Since the two corrections are completely different even in their sign, it is very interesting to use both in parallel for a mutual check. We applied this advantage to all measurements by choosing the time constants t_r lower than t_β and t_γ in our electronic counting chain I and higher (really the dead time in the coincidence channel, in order to avoid a big accidental correction) than the dead times in the single channels in the electronic apparatus II, both of which always operated in parallel. The results are very satisfactory, since even if the uncorrected decay rates differ by 3-4% the application of equations (6a) and (7a) leads to at least 99.9% agreement. This is the best confirmation of the correction formulae used, but still other checks were carried out. Fig. 9 shows the like-wise satisfactory results of varying the dead times and extrapolating to the dead-time zero, and Fig. 11 those obtained by varying the source strength. All the results confirm that the dead-time corrections used are correct to better than 0.1%.

7.5 — Decay-scheme-dependent corrections

The decay-scheme-dependent correction factor K_ε covers all count-rate-independent corrections which depend on the particular decay scheme concerned and are normally corrections to the γ-efficiency of the β-counter and to the conversion of γ-rays. The correction K_ε can be derived theoretically by consideration and calculation of all elementary processes in the source and the counter. But even then experimental determinations of the required constants are necessary and sometimes these cannot be measured with sufficient accuracy (e.g. the separation of Compton and photo-effects in the β-counter). We therefore prefered to determine the correction factor K_ε purely experimentally. This can be done by using an extrapolation technique, varying ε_β and extrapolating to $\varepsilon_\beta = 1$, where $K_\varepsilon = 1$. The problem is to vary the β-efficiency in such

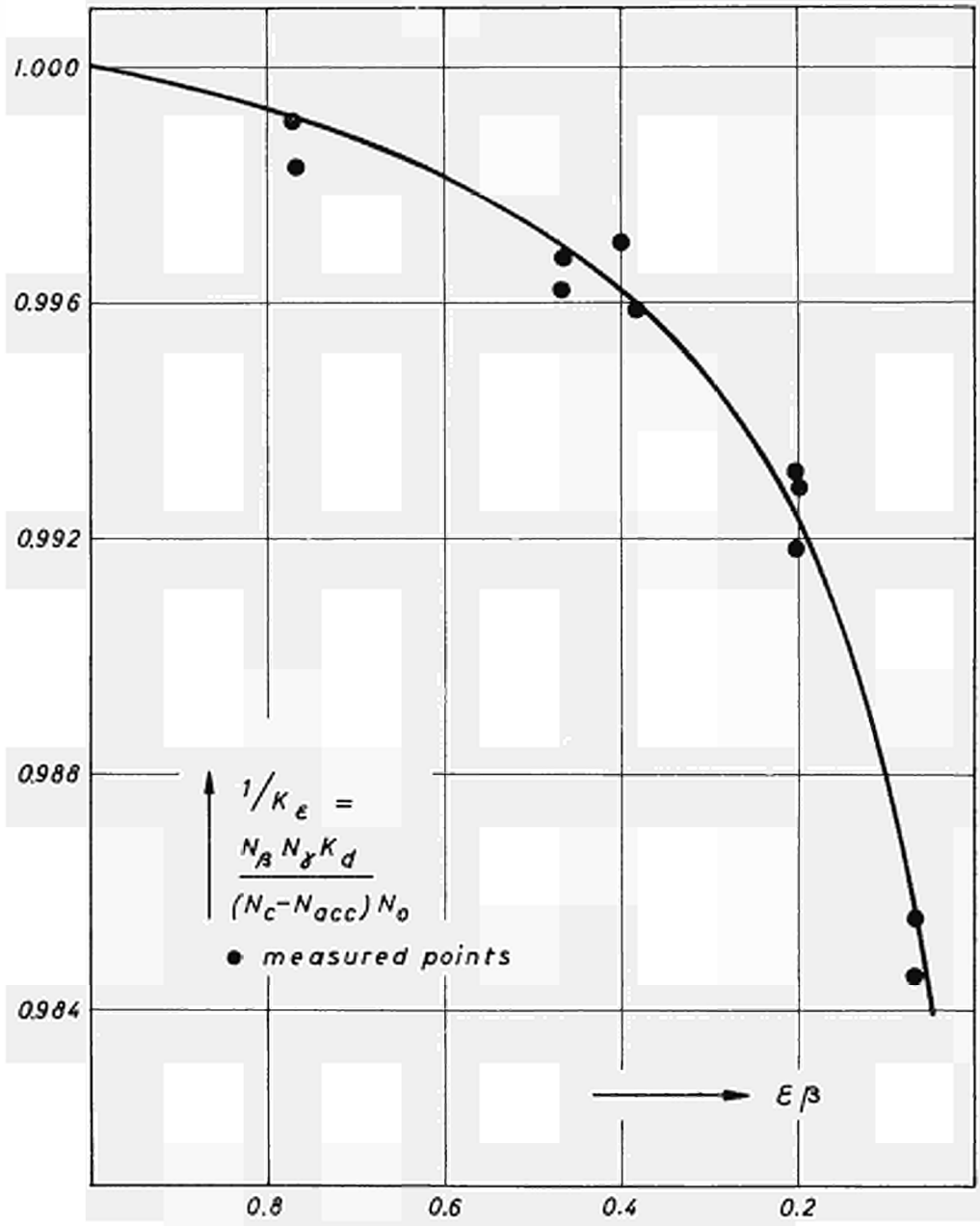

Fig. 12 — Measured values for K_ε as a function of ε_β

a way that all other conditions of the measurement remain unchanged. Covering the sources with supplementary foils seems to meet this requirement [9]. We therefore carried out such measurements and obtained the results shown in Fig. 12, which are at least $\pm 0.05\%$ accurate. Another method of varying the β-efficiency, that of varying the discrimination level in the β-channel, was also tried because of its simplicity. The results of these measurements agree with those discussed above only under certain conditions and are still then less satisfactory. Thus this method of varying the β-discrimination cannot be recommended.

It can be seen from Fig. 12 that K_ε is not proportional to $(1 - \varepsilon_\beta)/\varepsilon_\beta$ over the whole range of the β-efficiencies. This is in contradiction to the result of Williams and Campion [9] but may be attributable to the different discrimination in the γ-channels in their experiment.

8 — ERRORS IN OUR MEASUREMENT AND HOW THEY ARE STATED

The statement of errors in radioactive standardization is generally a rather arbitrary procedure. Even the definitions used are not always the same; consequently, comparison of the results of different working groups is usually very difficult if not impossible. The reason for this arbitrariness seems simply to be that the various individual errors are not really measured in a well-defined way but are estimated arbitrarily. In the Radio-isotopes Group of the CBNM we therefore always follow a definite procedure for the measurement and statement of the errors.

TABLE 5

Comparison of the various equations for the statistical error with the experimental standard deviation

No.	Source No.	Exp. st. dev.	Wolf (1) formula	C.T. (1) form.	No.	Source No.	Exp. st. dev.	Wolf form.	C.T. form.
1	4084	0.055	0.019	0.048	18	5056	0.088	0.053	0.084
2	4084	0.044	0.021	0.047	19	5056	0.102	0.054	0.084
3	5004	0.14	0.30	0.19	20	5056	0.136	0.051	0.083
4	5004	0.13	0.31	0.19	21	5056	0.090	0.051	0.093
5	5004	0.16	0.25	0.15	22	5057	0.081	0.054	0.085
6	5004	0.11	0.16	0.13	23	5057	0.078	0.055	0.086
7	5004	0.12	0.18	0.13	24	5057	0.099	0.047	0.076
8	5005	0.18	0.31	0.19	25	5057	0.084	0.055	0.095
9	5005	0.16	0.31	0.19	26	5075	0.096	0.58	0.081
10	5005	0.18	0.30	0.18	27	5075	0.108	0.62	0.127
11	5005	0.20	0.29	0.18	28	5078	0.134	0.049	0.106
12	5028	0.075	0.06	0.090	29	5078	0.109	0.046	0.072
13	5028	0.12	0.075	0.090	30	5081	0.063	0.034	0.098
14	5028	0.10	0.040	0.095	31	5081	0.081	0.036	0.099
15	5028	0.095	0.040	0.095	32	5082	0.092	0.038	0.085
16	5051	0.088	0.06	0.09	33	5082	0.306	0.015	0.086
17	5051	0.100	0.05	0.100					

(1) The formula given by Wolf [23] is $\sqrt{N_\gamma - N_c}/N_c$, that given by Campion and Taylor [18] $\sqrt{\{2 + (1 - \varepsilon_\beta - \varepsilon_\gamma)/\varepsilon_\beta\varepsilon_\gamma\}/N_0}$;
ε_γ was varied from 10 to 33%, ε_β from 40-98%.

First, the purely experimental standard deviation of the single measurement is determined for every source. (It is better to state the standard deviation of the single measurement because it is independent of the number of measurements and therefore more characteristic of the reproducibility.) Next, all possible individual errors, including those of the corrections, are measured

and their mean fluctuation is determined. These (standard deviations of the single measurements) are the basis of all statements of errors : in particular they are used for the statement of the final error of the measurement either by "statistical summation" according to the formula $\sqrt{\sum \Delta_i^2}$ or by arithmetical addition. This method of describing the errors is not entirely satisfactory, because errors can be forgotten and because the formula used for the "statistical summation" is only applicable if all variables associated with errors are factors in the final formula for the decay rate, which is not completely correct. It is however a well-defined method which anyone can check and reproduce and which can serve as a basis for comparisons of different measurements; consequently, we are using it until a better method is found. In our measurements the errors determined according to the principles discussed are as follows : $\pm 15\%$ in the dead-time correction (twice the measured mean fluctuation of the dead times involved); $\pm 10\%$ in the correction for accidentals (twice the measured mean fluctuation of the resolving time); $\pm 10\%$ in the γ-backgrounds (twice the measured mean fluctuation); $\pm 0.1\%$ in the decay rate for the decay-scheme-dependent correction K_ε (twice the standard deviation of the decay-rate measurements foi the determination of K_ε). An error in the β-background can be neglected. The statistical error was calculated from the Campion-Taylor formula [18] which was experimentally proved correct (table 5). A supplementary, somewhat arbitrary, error was introduced for the characterization of the stability of the counting electronics. It was simply taken as the half difference between the results for the decay rate of the two parallel equipments. The final errors for our measurements are then determined according to the statistical formula $\sqrt{\sum \Delta_i^2}$ from the individual errors Δ_i; this gave us $0.15-0.5\%$, which can be taken as the absolute accuracy of our measurements. In the course of the several hundred measurements performed during the last 4 years we have never found that the results of two different measurements did not agree within these limits.

9 — DETERMINATION OF THE "BEST" VALUE OF THE DECAY RATE AND TRACING OF FAULTS

If the decay rate is determined simply as the mean of the results of several measurements, numerous other experimental results obtained during the measurements are ignored. These supplementary experimental data not normally considered for the determination of the decay rate are : the efficiencies $\varepsilon_{\gamma I}$, $\varepsilon_{\gamma II}$ (and, for similar sources, also $\varepsilon_{\beta I}$ and $\varepsilon_{\beta II}$) and the ratios of the different results in the two counting chains N_{0I}/N_{0II}, $\varepsilon_{\gamma I}/\varepsilon_{\gamma II}$, $\varepsilon_{\beta I}/\varepsilon_{\beta II}$, $A = N_{\beta I}(1 - t_{\beta II}N'_{\beta II})/N_{\beta II}(1 - t_{\beta I}N'_{\beta I})$, $B = N_{\gamma I}(1 - t_{\gamma II}N'_{\gamma II})/N_{\gamma II}(1 - t_{\gamma I}N'_{\gamma I})$ and $C = (N_{cI} - N_{accI})\ (1 - t_{\beta II}N'_{\beta II})\ (1 - t_{\gamma II}N'_{\gamma II})\ K_{dII}/(N_{cII} - N_{accII})\ (1 - t_{\beta I}N'_{\beta I})\ (1 - t_{\gamma I}N'_{\gamma I})\ K_{dI}$. All these data were compiled in a special table, from which it could be seen that most of them were reproducible with the same accuracy as the decay-rate results. Thus we got, for example, the following mean results and standard deviations for a series of measurements carried out during November and December 1962 : $\varepsilon_{\gamma II} = (33.06 \pm 0.15)\%$, $\varepsilon_{\gamma I}/\varepsilon_{\gamma II} = 1.006 \pm 0.3\%$, $\varepsilon_{\beta II} = (97.5 \pm 0.2)\%$, $\varepsilon_{\beta I}/\varepsilon_{\beta II} = 1.000 \pm 0.2\%$, $N_{0I}/N_{0II} = 0.999 \pm 0.14\%$, $A = B = C = 1.000 \pm 0.15$ to 0.25%. In order to make full use of all these experimental data, a complicated procedure should be developed. This we have not yet done. We have however used the above-mentioned table for the rejection of all measurements in which one of the data exceeds the mean value by more than 3 times the mean fluctuation. Only the remaining results were used for the calculation of the mean value of the activity.

The above mentioned table of all experimental data can also be used for the detection of instrument defects. These are usually incorrect timing or faulty pulses in one channel (and sometimes loss of pulses in one channel). If one of the two timers is not working properly, the efficiencies and their ratios remain right but all other data differ from the mean values. (If only one timer were used for the two counting chains, timing errors would not be seen.) If faulty

pulses are counted in one channel of one counting chain, then the efficiency for this channel, the ratio of the efficiencies and the ratios of the N_0 and A or B or C will differ from the mean values. Thanks to such checks we have detected several faults of the order of 0.5% and less (e.g. malfunctioning of a timer), which would otherwise have been very difficult to find.

10 — CONCLUSIONS

The detailed investigation of the $4\pi\beta$-γ-method confirmed the result of Campion [1], thereby showing that this method allows absolute accuracies of 0.1% to be achieved in the determination of decay rates. This precision can only be obtained if the electronic circuitry is designed and adjusted very carefully and if continuous tests guarantee its stability. The controls and practice at the CBNM, which are described in the paper, furthermore ensure that the activity of the sources prepared for the BIPM is known with an accuracy comparable to that of the method.

Our thanks are due to Dr. Spaepen, Director of the CBNM, for his constant encouragement and help during this work, to the radio isotopes group of the BIPM — in particular Dr. Rytz — for suggesting the BIPM work, and to this group and the radio isotopes group of the NPL — especially Mr. Williams — for their very kind and useful co-operation. Several members of the radioisotopes group of the CBNM (in particular Mr. Vaninbroukx and Mr. van der Eijk) helped us considerably during this work.

REFERENCES

[1] P.J. CAMPION, *I.J.A.R.I.* **4**, 232, 1959.
[2] C.J. MALETSKOS and J.W. IRVINE, *Nucl.* **14** (4), 84, 1956.
[3] R.L. BLANCHARD, B. KAHN and R.D. BIRKHOFF, *Health Phys.* **2**, 246, 1960.
[4] W. PARKER, M. DE CROES and K. SEVIER, *Nucl. Instr. Meth.* 7, 22, 1960.
[5] J. BRYANT, *I.J.A.R.I.* **13**, 273, 1962.
[6] B. OWEN and R.A. LLOYD, *Nat.* **181**, 396, 1958.
[7] A.B. GILLESPIE, *Signal, Noise and Resolution*, Pergamon 1953.
[8] J. STEYN and J.F. HAASBROEK, Sec. Geneva Conf. 1958, Pap. 1104.
[9] A. WILLIAMS and P.J. CAMPION, *I.J.A.R.I.*, at press.
[10] E.R. MOSBURG and W.M. MURPHY, *React. Sc. Techn.* **14**, 25, 1962.
[11] J. THOMAS, RISÖ Rep. No. 57, 1963.
[12] W. PÖNITZ, Internal report Inst. f. angew. Kernphys. KFK. Karlsruhe, 1963.
[13] J. BRYANT, *I.J.A.R.I.* **14**, 143, 1963.
[14] P.J. CAMPION in *NBS Handbook* **80**, 1961.
[15] G.A. BRINKMAN, thesis, Amsterdam 1961.
[16] W.B. MANN and H.H. SEELIGER, *J. Res. Nat. Bur. Stand.* **57**, 257, 1956.
[17] R.D. EVANS, *The Atomic Nucleus*, Mc Graw-Hill, 1955.
[18] P.J. CAMPION and J.G.V. TAYLOR, *I.J.A.R.I.* **10**, 131, 1961.

Other fundamental references not separately mentioned are :

[19] J.L. PUTMAN, *A.E.R.E.* I/M **26**, 1957.
[20] K.P. MEYER, P. SCHMID and P. HUBER, *Helv. Phys. Acta* **32**, 423, 1959.
[21] A. GANDY, *Rev. Series I.A.E.A.* No **14**, 1961.
[22] F. BROWN and D.E. WATT, *AWRE* O-**28**/61, 1961.
[23] G. WOLF, *Nucleonik* **2**, 255, 1961.
[24] A. GANDY, *I.J.A.R.I.* **13**, 501, 1962.